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Progress Report of Disposal Area Studies at Oak Ridge National Laboratory: Period of October 1, 1975 to September 30, 1977

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ENVIRONMENTAL SCIENCES DIVISION Publication No. 1286

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OAK RIDGE NATIONAL LABORATORY

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NUCLEAR FUEL AND WASTE PROGRAMS

Environmental Methods for Management of Low-level Radioactive Waste (ONL-WLO9)

PROGRESS REPORT OF DISPOSAL AREA STUDIES AT OAK RIDGE NATIONAL LABORATORY: PERIOD OF OCTOBER 1, 1975 TO SEPTEMBER 30, 1977

T. Tamura, L. D. Eyman, A. M. Stueber, and D. S. Ward³

ENVIRONMENTAL SCIENCES DIVISION Publication No. 1286

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ABSTRACT

TAMURA, T., L. D. EYMAN, A. M. STUEBER, and D. S. WARD. 1980. Progress report of disposal area studies at Oak Ridge National Laboratory: Period of October 1, 1975 to September 30, 1977. ORNL/TM-5514. Oak Ridge National Laboratory, Oak Ridge, Tennessee. 92 pp.

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From October, 1975 to September, 1977 activities were undertaken to further understand or correct the release of radioactivity to the White Oak Creek watershed from burial grounds and other waste management facilities. Investigation of 60 Co migration from trenches 5 and 7 and pit 4 showed that between 90 and 95% of 60 Co is mobilized by the tetramethyl ester of EDTA, which is a common decontamination-process substance. Results from a kinetics study indicate 60 Co is taken up by either insoluble organics, iron, or manganese oxides. The observed manganese oxide-cobalt association was the greatest of the three, with a correlation of greater than 0.95.

Estimations of annual 90 Sr discharge into White Oak Creek from Solid Waste Disposal Area (SWDA) 4 were made using the difference between annual precipitation and evapotranspiration. Another method, stream-monitoring, gave higher values of 90 Sr discharge. Possibly, the discrepency was due to a malfuntioning monitoring station. However, unaccounted sources such as SWDA's 1 and 3, contaminated soils on the flood plain of the creek adjacent to SWDA 4, and along the creek channel above SWDA 4 may also have been responsible for the discrepency. Further study of these possible sources is necessary.

Corrective methods employed were: a surface diversion system in SWDA 4; the sealing of several trenches in SWDA 5; and the emplacement of bentonite surface seals in designated areas of the SWDAs.

A new technique for tracing ground-water movement and dating ground-water using halocarbons was developed and tested. Three tests were run in areas of well-known hydrology using tritium as a standard. In the first test the halocarbon did not indicate the presence of younger water at depth. However, in the other two the correlation between the halocarbon and tritium was good. The presence of a fault-zone was indicated by the halocarbon in one test. A final, practical test was carried out in the ground-water adjacent to SWDA 6. Good results were again obtained indicating the technique is viable and should be further developed.

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INTRODUCTION

The release of radioactivity from White Oak Creek Watershed to the Clinch River has steadily declined over the past years. This decline reflects both improvements in process waste treatment methods and remedial measures implemented in Solid Waste Disposal Areas (SWDA) as a result of studies carried out under this program. The continuing primary goal of the program is to further reduce or eliminate these releases in order to meet the Department of Energy's "as low as practicable" standards.

Several corrective measures were implemented during the reporting period in the SWDAs in order to decrease the release of radioactivity from these areas. These measures include the application of bentonite surface seals in specific areas of the SWDAs, installation of a surface water diversion system in SWDA 4 and the sealing of specific trenches in SWDA 5. Evaluation of the effectiveness of these measures is continuing and is an important factor in determining the engineering solution necessary to meet the primary goal of the program.

Preliminary results of a comprehensive sampling program above station 3 indicate that releases of 90 Sr which have been attributed to SWDA 4 may arise in part from additional sources along White Oak Creek. Some of these inputs of 90 Sr are relatively diffuse, and many indicate a relatively widespread source of 90 Sr to the creek along its course.

During FY-1977 a major recruiting program was completed with the addition of eight research staff members to the project. These individuals bring expertise to the project in the areas of hydrology,

hydrologic modeling, geochemistry, soil mechanics, soil physics, and stratigraphy. New initiatives in research within the above disciplines are getting underway.

SEEPAGE TRENCH 7

Mechanisms of 60Co Transport

The role of organics in 60 Co migration was investigated through the use of gel filtration chromatography (GFC). In addition to evaluating the roles of various organics in 60 Co mobilization, GFC permits differentiation between organic and inorganic modes of transport.

Chromatographic gels cause fractionation of organic compounds by molecular weight as described below, using Sephadex gel G-10 as an example. Gel G-10 is designed to retard the passage of organic solutes with molecular weights less than approximately 700, calibrated by dextrans. Organic solutes greater than 700 mol. wt., whether 750 or 2,000,000 are eluted first as the so-called void volume. Smaller molecules are retarded and fractionated so that the smallest species, usually inorganics, are the last to emerge from the column. Sephadex gels G-15 and G-25 operate according to the same principle, but their void volumes are 1500 and 5000 in mol. wt., respectively.

In the present investigation, 1-ml aliquots of concentrated ground water samples were eluted through gels G-10, G-15, and G-25. Upon elution from the column, organic carbon was measured on an in-line UV spectrophotemeter by absorbance at 280 nm. One- to 3-ml aliquots were

continuously collected for later 60 Co determinations. Elution for the GFC separations profiles display metal contents of the different molecular weight organic fractions (Figs. 1 and 2).

4

In all elution profiles in Fig. 1, three different organic fractions are seen. Ninety to 95% of the 60 Co is correlated with the middle fraction, which represents an organic with a molecular weight less than approximately 700. Five to 10% of the 60 Co is associated with the void volume, and no 60 Co is observed with the peak representing the smallest molecular weight organics. Elution profiles in Fig. 2, for a different ground-water sample, are almost identical to those of Fig. 1, the major difference being that two principal 60 Co-bearing fractions are observed.

Preliminary infrared spectroscopic evidence suggests that the large molecular-weight organics involved in minor 60 Co transport are humic substances from the soil. These are chemically similar to humic substances extracted from soils and shales of the study area. The major 60 Co-bearing organic fraction is likely a heterogenous group of synthetic compounds of similar molecular weight leached from the waste pits. These low molecular-weight compounds might include chelating agents such as EDTA and EDDA used in decontamination operations. However, our preliminary spectroscopic measurements on this fraction show no evidence for chelating compounds, implying that these are not major constituents.

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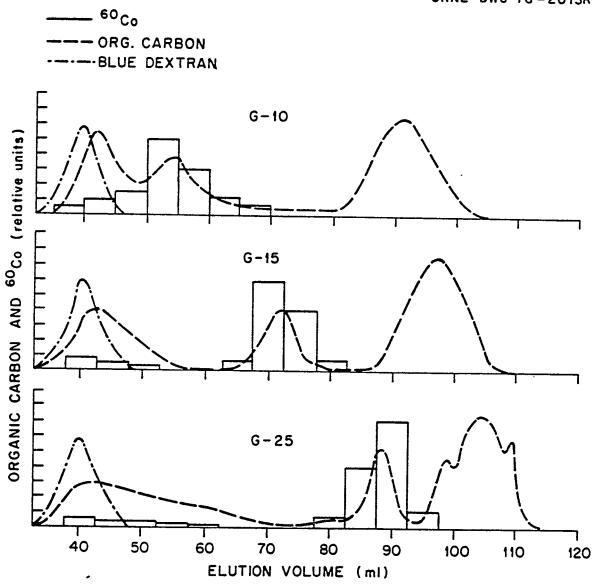


Fig. 1. GFC elution profiles of ground-water sample CS-1, collected from RS-7, east of Trench 7.

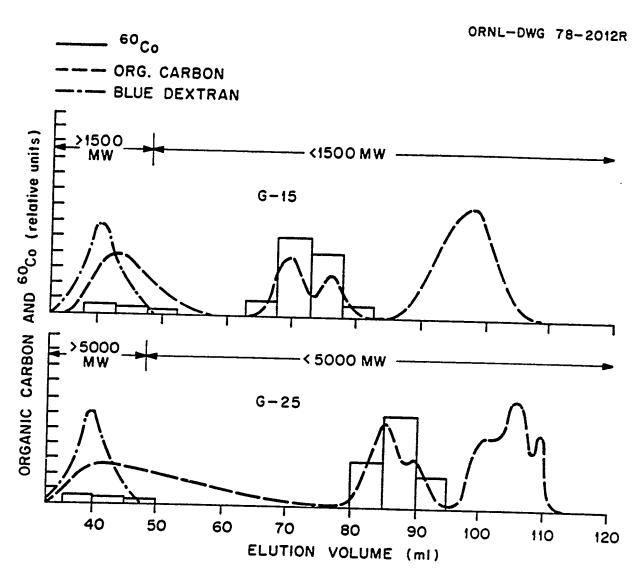


Fig. 2. GFC elution profile of ground-water sample MW, collected from a well 7.6 m east of Trench 7.

Our research in 1976 showed that the intermediate fraction transporting most 60 Co contains phthalate esters, which are common constituents of plastics. Phthalate esters and numerous related compounds, many of which are acidic, have also been identified in sanitary landfill leachates. The occurrence of these plastic degradation products appears to be widespread in ground waters near disposal areas.

Phthalate esters and related compounds may be involved in the transport of 60 Co in the ORNL disposal areas. If these compounds mobilize radionuclides, the problem is likely to be long-term in that degradation of plastics in the environment is very slow.

A compilation of selected radionuclide analyses for water samples collected from seeps adjacent to pit 4, trench 5, and trench 7 is given in Table 1. Sampling sites are indicated in Fig. 3.

Through the use of gel filtration chromatography (GFC) and infrared spectroscopy, we determined during 1977 that 5 to 10% of the 60 Co in seep waters in the vicinity of pit 4, trench 5, and trench 7 is complexed with humic substances derived from the soil. However, because ground water in and very close to the trenches is typically low in humic content, it is believed that soil organics are not major contributors to radionuclide dispersion from the trenches. Instead, humics become associated with radionuclides some distance from the trenches, particularly in the seeps, where ground-water humic concentrations are the greatest.

Cobalt-60 analyses of 0.22 micron-filtered water samples from selected seeps adjacent to Pit 4, Trench 5, and Trench 7. Table 1.

| | ··· | |
|--------------|---------|----------|
| Well code | Date | (pCi/ml) |
| RS3 | 6/24/75 | 41.8 |
| RS5 | 6/25/75 | 17.6 |
| RS7 | 6/26/74 | 301.4 |
| | 7/12/74 | 232.0 |
| | 8/15/74 | 241.0 |
| | 6/23/75 | 190.1 |
| | 7/20/76 | 81.1 |
| RS9 | 6/24/75 | 36.4 |
| | | |

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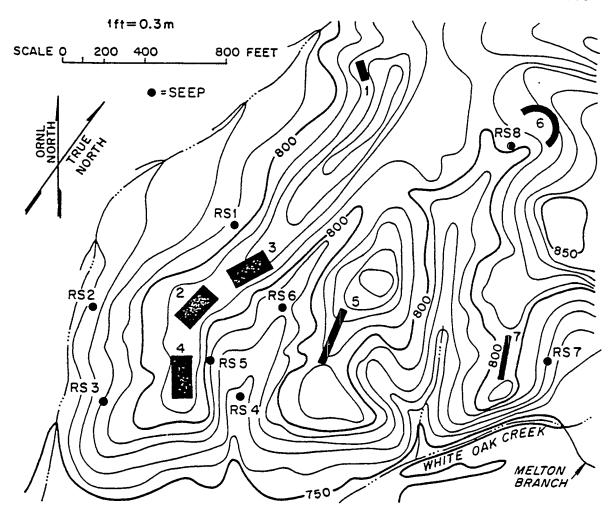


Fig. 3. Location of seeps associated with pits 1, 2, 3, and 4, and trenches 5, 6, and 7 in ORNL disposal areas.

Our GFC analyses have also indicated that the majority of the $^{60}\mathrm{Co}$ present in ground water from the trench 7 area is complexed with a strong ligand having a molecular weight of approximately 300. Recently this ligand has been unequivocally identified. Concentrated and purified ground water was methylated with ${\rm BF_3\text{-}MeOH}$ and analyzed by gas chromatography-mass spectrometry (GC-MS), using a procedure adapted from Rudling (1972). The GC profile for methylated ground water is illustrated in Fig. 4. The dominant peak represents the tetramethyl ester of EDTA (ethylenediaminetetraacetic acid), an extremely strong chelate commonly employed in decontamination operations at nuclear facilities. The mass spectrum of the tetramethyl ester of EDTA is exhibited in Fig. 5. High resolution MS verifies the EDTA identification through the elemental composition of the mass peak and fragments. Through use of CDTA (cyclohexanediamine-tetraacetic acid) as an internal standard, the EDTA concentration of this sample has been calculated to be approximately 3.4 \times 10⁻⁷ M. EDTA has also been detected in samples obtained near pit 4 and trench 5.

Other organic acids identified include palmitic acid and phthalic acid (in a different sample) which are much weaker complexing agents than EDTA. Strong chelates similar to EDTA, such as NTA (nitrilotriacetic acid) and diethylenetriamine-pentaacetic acid (DTPA) are below the detection limits of this analysis, which is approximately $3.0 \times 10^{-9} \, \underline{\text{M}}$. NTA is biodegradable (Swisher et al., 1974) and would not be expected in significant concentrations in the ground water even if it had been originally present in the waste. DTPA and other

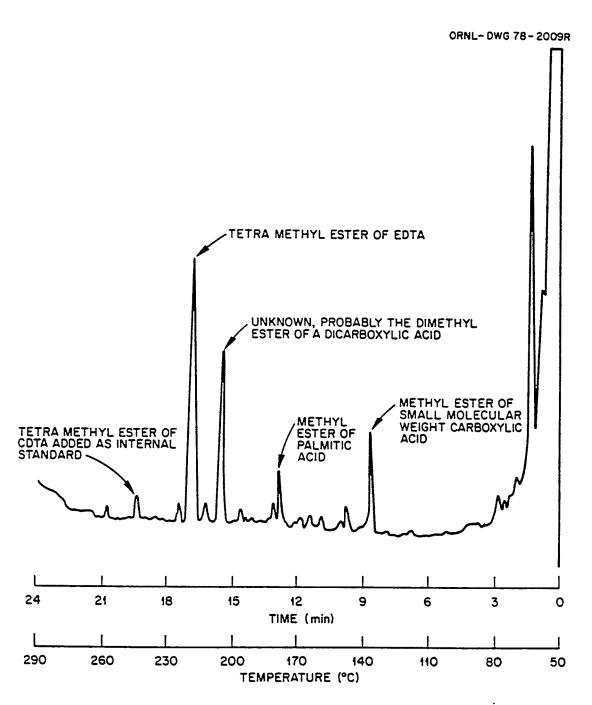
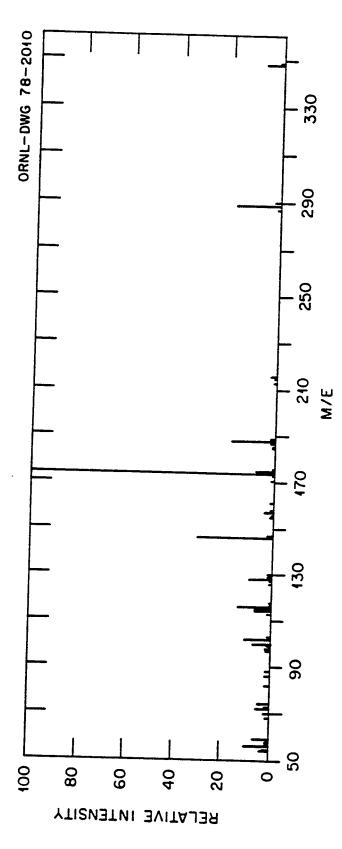


Fig. 4. GC profile of GFC-purified and methylated ground water from cobalt seep.

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Mass spectrum of tetramethyl ester of EDTA, from GFC purified and methylated ground water from cobalt seep. Fig. 5.

multidentate chelates were used only sparingly in decontamination at ORNL during the 1950s and 1960s and consequently do not appear to be significant in mobilization at this site.

EDTA is therefore reasoned to be the dominant mobilizing agent for 60 Co in ground water from the vicinity of trench 7 and in samples from the vicinity of pit 4 and trench 4.

Adsorption Kinetics of ⁶⁰Co

The ⁶⁰Co contents of different particle size fractions of soil from the trench 7 seep were first evaluated. If ⁶⁰Co was adsorbed by clays, the less than 2-µm fraction should contain essentially all of the radionuclide. If the adsorbents were particle coatings such as Fe and Mn oxides or insoluble organic matter, all of which are known to have strong sorption capabilities for ⁶⁰Co and various transuranics (Jenne, 1968; Burns, 1976; Kunzendorf and Friedrich, 1976), ⁶⁰Co should be observed in all size fractions. However, the fines would possess the highest radionuclide concentrations because of their greater particle surface area, and therefore the greater amounts of particle coatings available for adsorption. The data (Fig. 6) point to adsorption by Fe and Mn oxides or insoluble organics rather than clays.

Next we attempted to differentiate the relative roles played by Fe oxides, Mn oxides, and insoluble organics in 60 Co sorption. The first approach was to correlate total free Mn oxide, free Fe oxyhydroxide, and organic carbon content with 60 Co concentrations in these size-fractionated soils and in other unfractionated samples and sediments from the trench 7 area. Correlation coefficients for

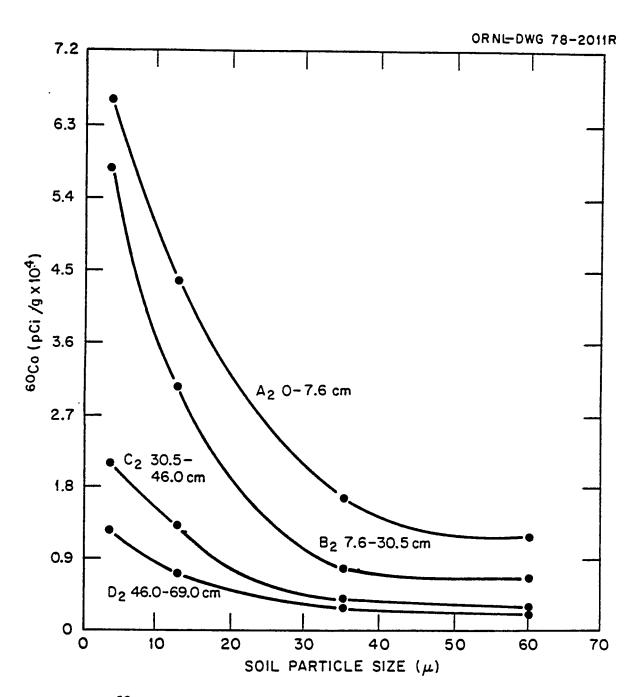


Fig. 6. ^{60}Co versus particle size for four soil samples taken from different depths from cobalt seep east of Trench 7.

 60 Co-Mn were highest, generally greater than + 0.8, indicating a high degree of association between the two factors. The 60 Co-Fe correlation coefficients averaged approximately + 0.5, suggesting a possible weak association, and 60 Co-organic carbon demonstrated a complete lack of correlation.

The partitioning of 60 Co between Mn and Fe oxides was further investigated through wet chemical extractions, using acidified hydroxylamine hydrochloride and acidified ammonium oxalate for the relatively selective dissolution of the Mn oxides and amorphous Fe oxyhydroxides, respectively. Soil samples were extracted in succession with the two reagents, and extractants were analyzed for Mn, Fe, and radionuclide. Correlation coefficients for 60 Co-Mn and 60 Co-Fe leave no doubt that 60 Co is primarily associated with Mn oxide phase. Figure 7 illustrates the 60 Co and Fe contents of 96 different extracts on soils from the cobalt seep east of trench 7. Data points are extremely scattered, and no correlation is observed. On the other hand, 60 Co and Mn concentrations in extracts from the same soils (Fig. 8) are related with correlation coefficient of 0.9570, indicating an extremely high degree of association between Co and Mn.

SOLID WASTE DISPOSAL AREA 4

Discharge of 90Sr

Solid Waste Disposal Area 4 is located approximately 0.8 km southwest of ORNL in the lower portion of a small watershed (Figs. 9 and 10). The discharge of 90 Sr from this burial ground to White Oak Creek cannot be measured directly. It has been estimated in the past

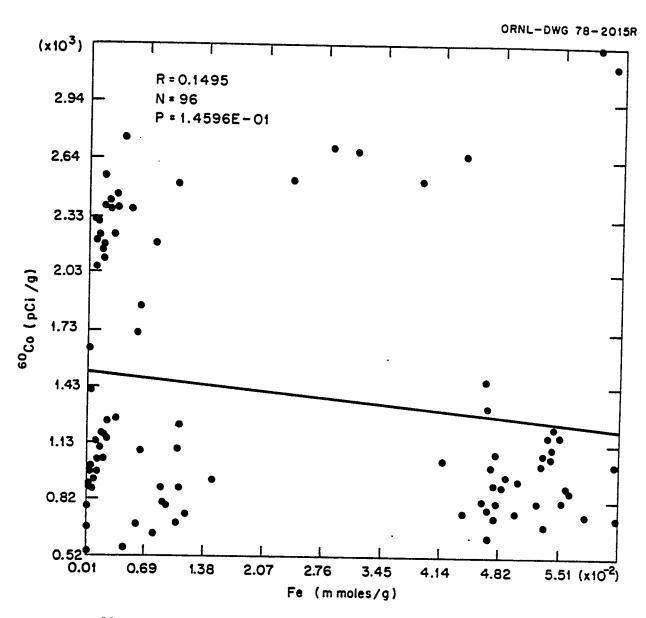


Fig. 7. ⁶⁰Co and Fe contents of 96 extracts of soil from the cobalt seep, with best fit regression line and correlation coefficient.

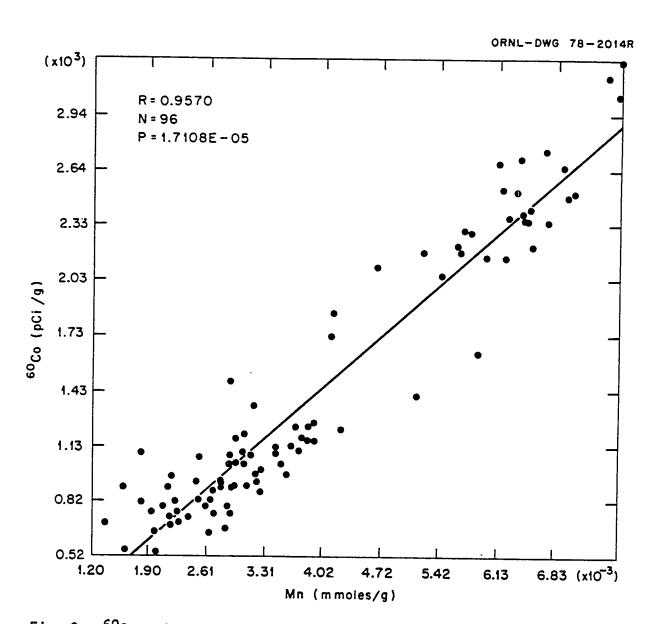
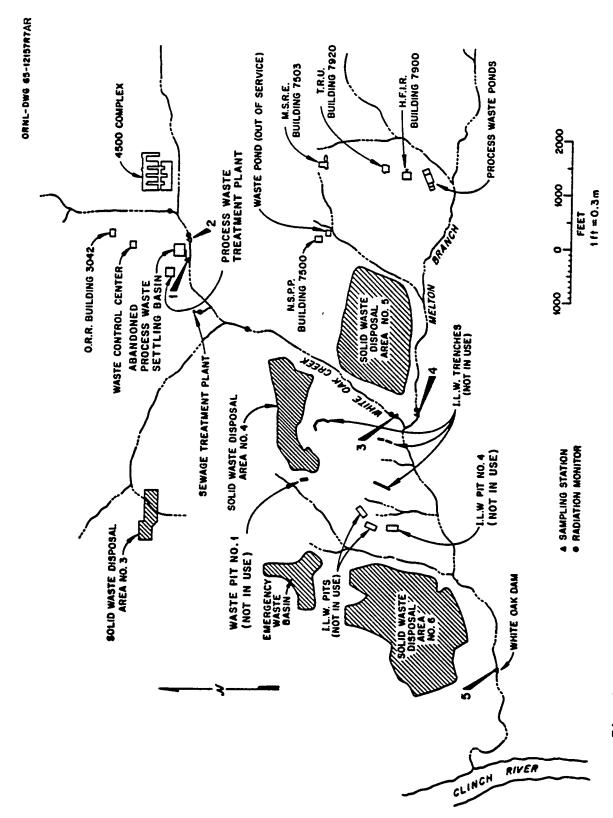


Fig. 8. ^{60}Co and Mn contents of 96 extracts of soil from the cobalt seep, with best-fit regression line and correlation coefficient.

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Approximate location of disposal areas and sampling stations at ORNL. 6 Fig.

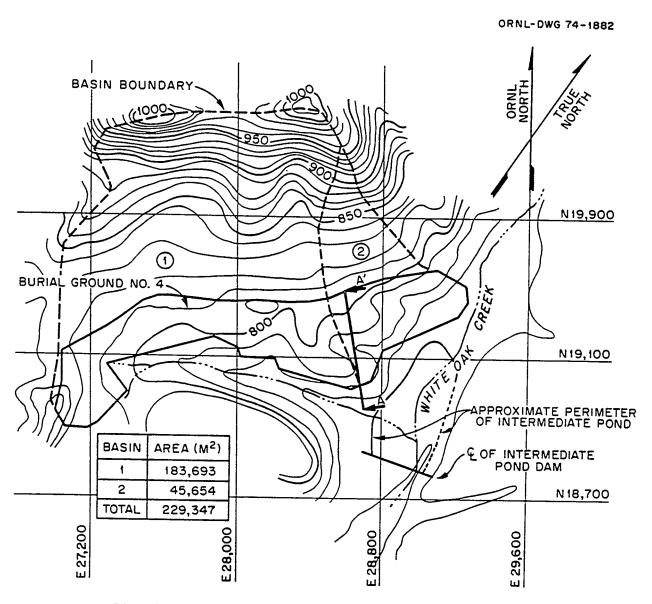


Fig. 10. Location of SWDA-4 within small watershed.

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through the use of a network of wells and surface-sampling sites established adjacent to the disposal area on the floodplain of White Oak Creek (Duguid, 1975). These locations (Fig. 11) have been sampled approximately three times a year and the average concentration of 90 Sr in water entering White Oak Creek from the disposal area has been estimated from these samples. The total annual discharge of 90 Sr from SWDA 4 has in turn been obtained by multiplying the average 90 Sr concentration by a flux of infiltrated water that has been estimated according to a technique based upon the difference between annual precipitation and evapotranspiration values for Walker Branch Watershed (Duguid, 1975).

The apparent annual ⁹⁰Sr discharge from SWDA 4 can also be obtained from stream-monitoring data along White Oak Creek, by subtracting the ⁹⁰Sr discharged from plant operations (stations 1 and 2) from the ⁹⁰Sr discharged at monitoring station 3 (Fig. 9). The difference includes any discharge from several other possible sources, which have been regarded as insignificant since a reconnaissance sampling of them was carried out in 1973 (Duguid, 1975). The discharge of ⁹⁰Sr that may be attributed to SWDA 4 on the basis of stream-monitoring data for water years 1963 through 1977 is presented in Table 2.

The average concentration of 90 Sr in the water flowing from SWDA 4 during water year 1977, obtained from sampling of the monitoring network in October 1976 and in May 1977, is shown in Table 3, along

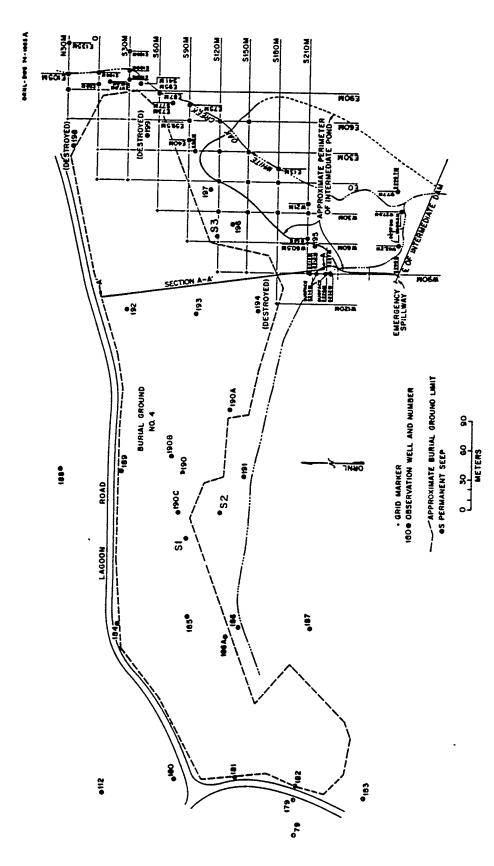


Fig. 11. Location of ground-water monitoring wells near SWDA-4.

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Table 2. Discharge of 90Sr from SWDA-4 and precipitation data for water years 1963 through 1977

| Water year ^a | Precipitation (cm) | Total ⁹⁰ Sr discharge (Ci) | Discharge of ⁹⁰ Sr (mCi/cm) |
|----------------------------|--------------------|---|--|
| 1963 | 140.5 | 4.82 | 34.3 |
| 1964 | 106.9 | 2.98 | 27.9 |
| 1965 | 132.0 | 3.10 | 23.5 |
| 1966 | 103.8 | 2.52 | 24.3 |
| 1967 | 153.8 | 2.72 | 17.7 |
| 1968 | 114.3 | 2.04 | 17.8 |
| 1969 | 101.8 | 2.08 | 20.4 |
| 1970 | 121.7 | 1.60 | 13.1 |
| 1971 | 122.6 | 1.18 | 9.6 |
| 1972 | 120.4 | 2.36 | 19.6 |
| 1973 | 181.0 | 1.58 | 8.7 |
| 1974 | 174.7 | 5.22 | 29.9 |
| 1975 | 146.6 | 3.22 | 21.9 |
| 1976 | 123.9 | 5.12 | 41.3 |
| 1977 | 129.1 | 2.27 | 17.6 |

^aWater year is September 1 through August 31.

Table 3. Concentration ($\mu\text{Ci/ml}$) of ^{90}Sr flowing from SWDA-4

| Water year | Basin 1 | Basin 2 |
|------------|------------------------|------------------------|
| 1974 | 6.8 x 10 ⁻⁶ | 4.7 x 10 ⁻⁶ |
| 1975 | 6.4×10^{-6} | 3.9 x 10 ⁻⁶ |
| 1976 | 5.5×10^{-6} | 3.7×10^{-6} |
| 1977 | 8.3 x 10 ⁻⁶ | 6.1×10^{-6} |

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with the corresponding values for previous water years extending back to the time when the monitoring network was established. Calculation of the 90 Sr discharge from SWDA 4 according to the method of Duguid (1975), using the precipitation value shown in Table 2, yields an estimate of 1.1 Ci for water year 1977. Calculated estimates for the past five water years are compared with 90 Sr discharges based on stream-monitoring data in Table 4. With the exception of water year 1973, stream-monitoring indicates vastly greater 90 Sr discharge from SWDA 4 than that which is estimated by calculation. The discrepancies may indicate: (1) a malfunction of monitoring station 3; (2) inadequacies in the technique used to calculate 90 Sr discharge from SWDA 4; and/or (3) a new source of 90 Sr input to White Oak Creek above monitoring station 3.

It has been suggested (Duguid, 1976) that the higher recent $^{90}\mathrm{Sr}$ discharges from SWDA 4 that are indicated by stream-monitoring data could be due to a malfunction in monitoring station 3, which occurred post-1974 but was not discovered until 1976. While such a malfunction could have affected the apparent $^{90}\mathrm{Sr}$ discharge for water years 1975 and 1976, the discrepancies between calculated and stream-monitored discharges are not confined to these water years. Although the possibility exists that stream monitoring of $^{90}\mathrm{Sr}$ discharge in White Oak Creek may be less than satisfactory, such limitations should be applied to all the data; other possible reasons for the recent apparently higher discharges are being sought.

The technique used to calculate the annual 90 Sr discharge from SWDA 4 should be regarded as yielding only a crude estimate (Duguid, 1976). The flux of infiltrated water is obtained by neglecting the

Table 4. Total 90Sr discharge from SWDA-4 (Ci/year)

| Water year | Calculated | Stream monitoring |
|------------|------------|-------------------|
| 1973 | 1.7 | 1.58 |
| 1974 | 1.6 | 5.22 |
| 1975 | 1.2 | 3.22 |
| 1976 | 0.65 | 5.12 |
| 1977 | 1.1 | 2.27 |

surface runoff component of precipitation; however, any error introduced by this approximation will yield an apparent 90 Sr discharge from below the ground surface which is too high. A more significant potential source of error introduced by neglecting surface runoff lies in the possibility that 90 Sr is being leached from surficial sources and carried directly into White Oak Creek without being intercepted by the monitoring network. Furthermore, the average concentration of 90 Sr in water entering White Oak Creek from SWDA 4 is based upon samples from the monitoring network which are obtained only twice during the water year; the accuracy of sampling for a yearly average needs to be evaluated. In addition, the wells that comprise the monitoring network are very shallow and do not intercept any deep ground-water component that may have a different 90 Sr activity.

Possible Alternative Sources of 90Sr

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There is a possibility that SWDA 4 is not the only significant source of 90 Sr discharge to White Oak Creek between monitoring stations 2 and 3. Monitoring wells in the vicinity of disposal areas 1 and 3 and on the west side of SWDA 5 were last tested in 1973 (Duguid, 1975). Although at that time the 90 Sr activities in the water samples indicated that these potential sources were insignificant contributors to White Oak Creek, these monitoring wells should be checked again. They were sampled only one time during the reconnaissance survey; the 90 Sr activities in the ground waters may be a function of hydrologic parameters which vary with time.

Another potentially significant source of ⁹⁰Sr in White Oak Creek between monitoring stations 2 and 3 may be found in the soil on the floodplain of the creek adjacent to SWDA 4, and in the soil and sediment along the creek channel above SWDA 4. There is some evidence that suggests such a source may be responsible for the abnormally high 90 Sr discharge of White Oak Creek between monitoring stations 2 and 3 during recent water years. Binford and Gissel (internal communication, 1975) observed a linear relationship between White Oak Creek monthly flow and apparent $^{90}\mathrm{Sr}$ monthly contribution to White Oak Creek from SWDA 4 for the period June 1971 through March 1974 (Fig. 12). More recent data have been plotted to show similar linear relationships for the individual water years 1974, 1975, and 1976 (Figs. 13, 14, and 15). Such linear relationships imply that the 90 Sr concentration in White Oak Creek between monitoring stations 2 and 3 is essentially constant. Because monthly stream flow varies to some degree as a function of monthly rainfall, there must be a relatively rapid interaction between rainfall and the $^{90}\mathrm{Sr}$ source in order to maintain the relatively constant 90Sr concentration in the creek. This requirement seems to eliminate infiltration and movement through SWDA 4 and suggests a surficial source near White Oak Creek. Duguid (1976) reported significant 90Sr concentrations in the soils on the floodplain adjacent to the disposal area. The soil and sediment in the creek channel above the disposal area may be contaminated with 90sr

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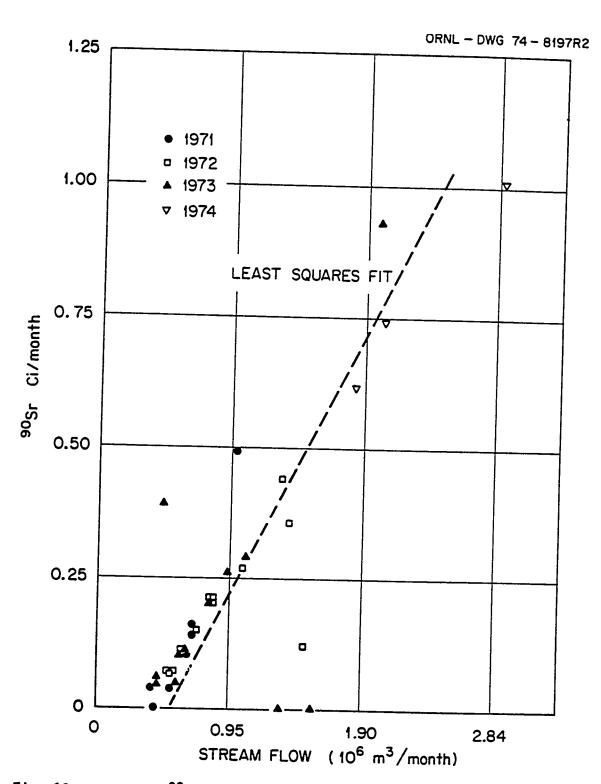


Fig. 12. Apparent 90Sr concentration to White Oak Creek from SWDA-4 (June 1971-March 1974).

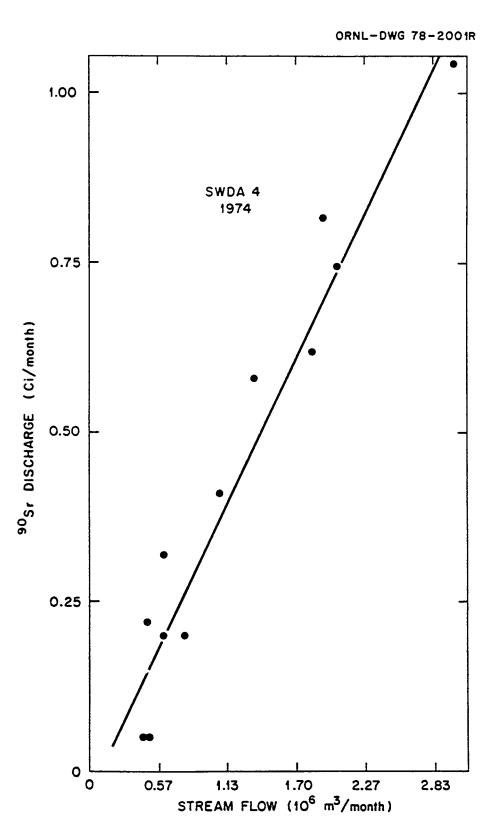


Fig. 13. Apparent 90 Sr concentration to White Oak Creek from SWDA-4 (1974).

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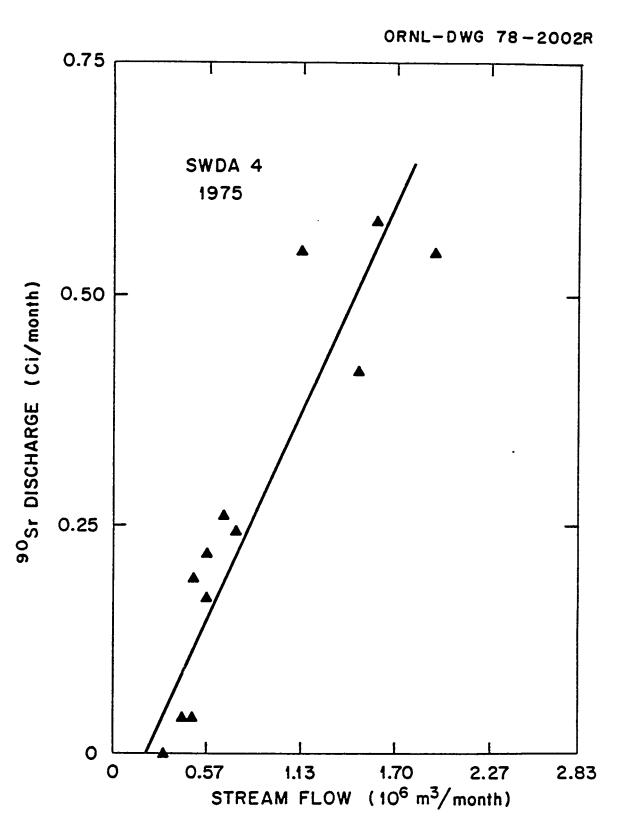


Fig. 14. Apparent $90 \mathrm{Sr}$ concentration to White Oak Creek from SWDA-4 (1975).

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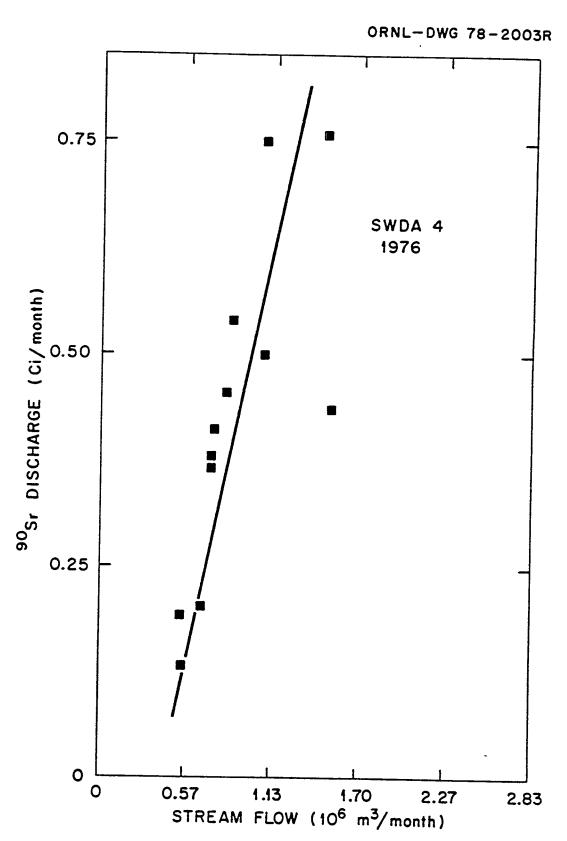


Fig. 15. Apparent $^{90}\mathrm{Sr}$ concentration to White Oak Creek from SWDA-4 (1976).

discharged from the Process Waste Treatment Plant. These potential surficial sources of $^{90}\mathrm{Sr}$ are being investigated further.

Soil Contamination

Soil cores collected from the floodplain below SWDA 4 were divided into subsamples and analyzed for 90 Sr and extractable strontium. The area sampled comprised an abandoned settling pond which was intermediate between the main Laboratory area and White Oak Lake. It was functional for only a short period during the 1940s. The purpose for obtaining the 90 Sr and extractable strontium data from this area is to determine the amounts of 90 Sr in the floodplain soils which may eventually be leached and transported into White Oak Creek.

Soil cores were collected to a depth of 17 to 35 cm and were sectioned into 2 or 3 subsamples depending on the core length. Each section was sampled and the sample was leached with hot concentrated HNO_3 for total ^{90}Sr determinations. Extractable Sr was determined by shaking 5 g of soil with 25 ml of 1 \underline{N} HCl or 0.5 \underline{M} CaCl₂.

The data indicate that soils from the floodplain contain between 222 and 1780 dpm/g of 90 Sr to a depth of at least 32 cm (Table 5). There were no "hot spots" identified in the survey, indicating a rather uniform distribution of 90 Sr in the area. However, higher concentrations of 90 Sr were found near the dam of the intermediate pond. This could represent either deposition of contaminated sediment during the pond operation or deposition of 90 Sr from SWDA 4 that passes through this area via an intermittent stream, or both. The data also indicate that deeper sampling will be required before an estimate

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Table 5. Strontium-90, total strontium, and total strontium extractable with CaCl₂ from soil samples collected below SWDA-4

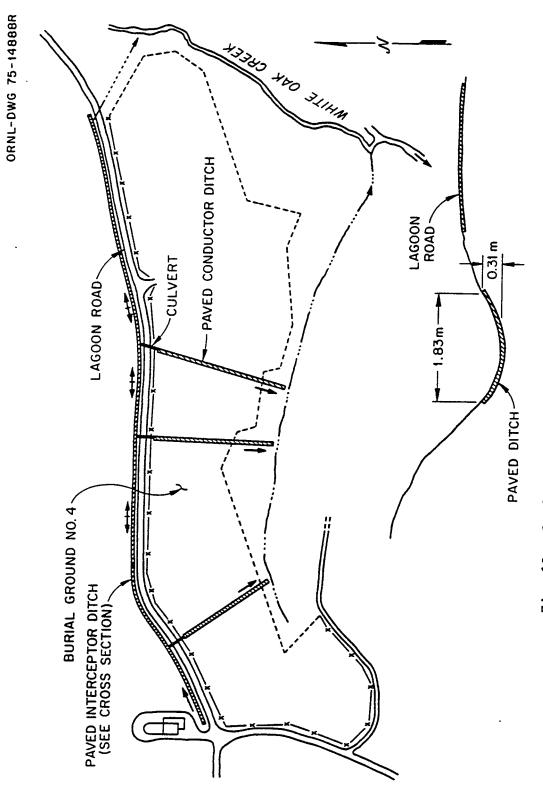
| Location | Depth (cm) | Total 90Sr (pCi/g) | Total Sr (1 <u>N</u> HC1) (ug/g) | Total Sr (extracted with 0.5 M CaCl ₂) (µg/g) |
|-----------|------------------------------|--------------------------|---|---|
| E60/S120 | 0-7 0-17 | 363 802 | 17.6 21.8 | 3.2 2.9 |
| E30/S210 | 0-7 0-17 | 141 100 | 4.9 3.8 | - <u>-</u> 2.4 |
| E30/S120 | 0-7 0-17 | 214 281 | 15.9 18.3 | 5.2 4.2 |
| E0/\$120 | 0 -7 7 - 22 | 113 113 | 10.6 6.0 | 3.5 3.9 |
| E0/S150 | 0-7 7-17 17-32 | 473 126 152 | 33.5 12.4 4.9 | 8.5 3.9 |
| E0/S180 | 0-7 7-22 | 194 236 | 15.5 16.5 | 4.3 4.3 |
| E0/S210 | 0-7 7-22 | 252 259 | 16.9 17.4 | 5.8 |
| E0/S240 | 0-7 7-17 | 118 118 | 8.9 5.0 | 4.4 |
| E0/\$270 | 0-7 7-22 | 405 468 | 19.6 21.7 | 5.2 |
| W30/S120 | 0-7 | 313 | | |
| W30/S120 | 0-7 7 - 22 | 281 | 11.0 9.2 | 6.2 6.7 |
| H30/S240 | 0-7 7-17 | 220 265 | 11.8 14.6 | 5.9 |
| i30/S270 | 0-7 7-22 | 477 513 | 13.4 11.5 | 7.4 |
| 130/5300 | 0-7 0-17 | 545 345 | 13.5 20.9 | 4.7 |
| 160/S240 | 0-7 7-17 | 554 416 | 7.6 5.0 | |
| 160/S270 | 0-7 7-17 | 545 671 | 10.6 9.7 | 10.5 |
| 160/\$300 | 0-7 7-17 | 793 761 | 13.0 10.7 | 9.2 8.2 |

of the total amount of $^{90}\mathrm{Sr}$ contained in the floodplain sediment can be made.

Although 90 Sr was not determined in soil extractants, considerable information may be gained from the total Sr analyses (stable Sr plus 90 Sr). Strontium extracted from soil with CaCl_2 solution would be readily exchangeable with soil water. Strontium extracted by 1 N HCL should be indicative of total Sr in the soil. On this basis 25 to 75% of the total Sr is potentially exchangeable with soil water. These results indicate that some 90 Sr will continue to discharge at a very slow rate to White Oak Creek even after the discharge from SWDA 4 has been reduced by restricting the amount of water moving through the disposal area.

Evaluation of Corrective Measures

Surface runoff and ground water from the drainage basin above SWDA 4 flow southward across the disposal area (Fig. 10). A suggestion had been made to construct a ground-water diversion trench along the north side of the disposal area. Because the estimated construction cost was prohibitively high, a surface runoff collector and diversion system was proposed, designed and constructed instead. The surface runoff collector consists of a shallow, asphalt-lined ditch along the north side of Lagoon Road above SWDA 4 (Fig. 16). The surface runoff is diverted across the disposal area by means of three asphalt-lined conductor ditches and one unlined natural drainage at the northeast edge of the disposal area.



Surface runoff diversion system in SWDA-4. Fig. 16.

Since the completion of the diversion system in September 1975, it has been observed to carry large amounts of water during heavy rains and small amounts of water for several days after a period of heavy rain. However, this corrective measure has had no apparent effect on the discharge of 90 Sr from SWDA 4. The average concentration of 90 Sr in water flowing from the disposal area has not decreased during water years 1976 and 1977 (Table 3).

A more specific evaluation of the diversion system can be obtained 90 Sr analyses of the surface stream along the south boundary of the disposal area, into which the conductor trenches empty (Fig. 16). Two branches of this stream have been sampled regularly as a part of the monitoring network (Fig. 11). The 90 Sr concentrations in the surface stream show no significant decrease since the installation of the diversion system (Table 6).

SOLID WASTE DISPOSAL AREA 5

Discharge of 90Sr

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Stream monitoring station 4 on Melton Branch provides data on radionuclide discharges to Melton Branch upstream from its confluence with White Oak Creek. Approximately 90% of the radioactivity measured at this station is attributed to SWDA 5, and the remaining 10% is from other sources within the drainage. The ⁹⁰Sr discharge from SWDA 5 and other sources, for water years 1967 through 1977, is presented in Table 7.

Table 6. Strontium-90 concentration (pCi/ml) in surface stream south of SWDA-4 $\,$

| Date | Surface stream #1 S214/W90 | Surface stream #2 S228/W90 | Average |
|-------|-------------------------------|-------------------------------|---------|
| 12/73 | 7.9 | 7.6 | 7.7 |
| 1/74 | 9.1 | 8.6 | 8.9 |
| 9/74 | 12.0 | 5.3 | 8.6 |
| | (Diversion | system installed - 9/75) | |
| 4/76 | 8.0 | 3.2 | 5.6 |
| 10/76 | 10.0 | 8.6 | 9.3 |
| 2/77 | 8.0 | 7.9 | 8.0 |
| 10/77 | 6.7 | 7.8 | 7.3 |

Table 7. Discharge of $90\mathrm{Sr}$ from SWDA-5 and precipitation data for water years 1967 through 1977

| Water year ^a | Precipitation (cm) | Total ⁹⁰ Sr Discharge (Ci) | Discharge of ⁹⁰ Sr (mCi/cm) |
|----------------------------|--------------------|---|--|
| 1967 | 153.8 | 0.89 | 5.8 |
| 1968 | 114.3 | 2.84 | 24.8 |
| 1969 | 101.8 | 0.88 | 8.7 |
| 1970 | 121.7 | 0.93 | 7.6 |
| 1971 | 122.6 | 0.58 | 4.7 |
| 1972 | 120.4 | 0.81 | 6.7 |
| 1973 | 181.0 | 1.43 | 7.9 |
| 1974 | 174.7 | 1.39 | 7.9 |
| 1975 | 146.6 | 2.07 | 14.1 |
| 1976 | 130.0 | 0.75 | 5.8 |
| 1977 | 129.1 | 0.43 | 3.4 |

aWater year is September 1 through August 31.

<u>Installation of Corrective Measures</u>

Significant concentration levels of radionuclides ²³⁸Pu, ²⁴⁴Cm, and ⁹⁰Sr had been discovered in the effluent from a small seep associated with trench 83 in SWDA 5. Beginning in May 1975, actions were taken to eliminate this discharge. Approximately 0.61 m of overburden were stripped from the area overlying trenches 105, 83, 72, and 69 (Fig. 17). Two underground dams were placed across trenches 83 and 105 (Duguid, 1976). The stripped area was then covered with a PVC membrane and the overburden was replaced. Grass was then planted in the area to prevent erosion. The operation was completed in September 1975, at the beginning of water year 1976. Therefore, the ⁹⁰Sr discharge data for water years 1976 and 1977 include the discharge from SWDA 5 after the corrective measures were completed.

Installation of a near-surface seal consisting of a bentonite-shale mixture was completed in October 1977 for 14 trenches in SWDA 5 (Fig. 18). The effectiveness in reducing infiltration of a bentonite application rate of 0.04 kg/m 2 over the 1765 m 2 area has not yet been determined.

The discharge of ⁹⁰Sr from SWDA 5 has been reduced significantly during the past two water years (Table 7); the data for 1977 represent the lowest values yet recorded. Undoubtedly the reduction is a result of the corrective measures described above, as well as others carried out concurrently (filling in of collapsed trenches, installation of concrete drainage ditches, and surface contouring for better drainage).

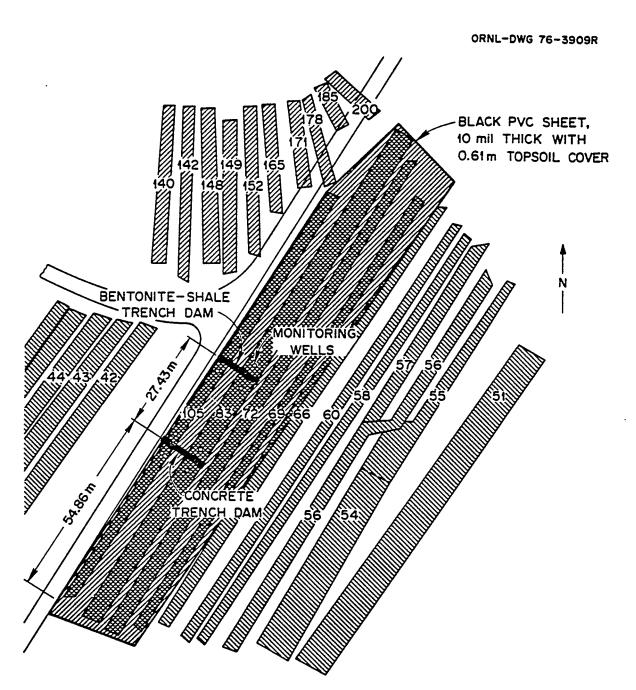


Fig. 17. Near-surface sealing of trenches in SWDA-5.

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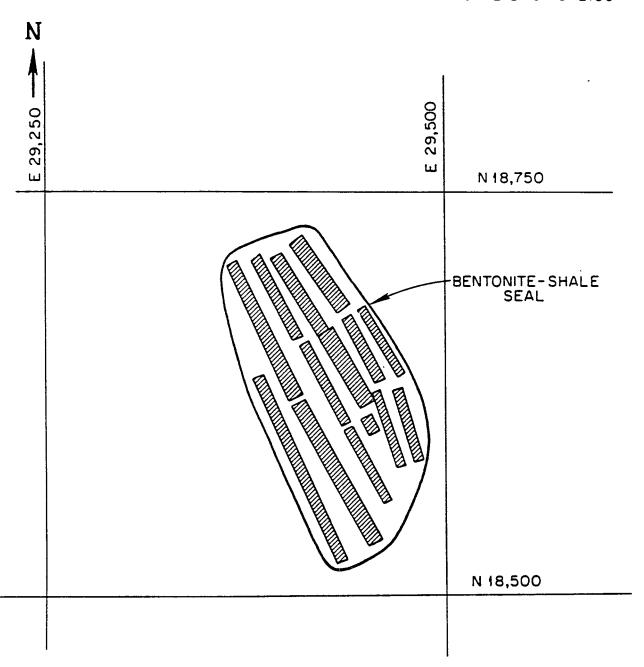


Fig. 18. Bentonite-shale surface seal in SWDA-5.

SOLID WASTE DISPOSAL AREA 6

Ground Water Contamination

During the wet winter months water infiltrates into the waste disposal trenches faster than it seeps out of the trench bottom. This causes water to accumulate in the trench and subsequently increases the leaching of the waste. For future studies of radionuclide transport from the disposal areas located on the Oak Ridge reservation it is important to have concentration data for radionuclides in the water within the disposal trenches. Thus, during the spring of 1976 water samples were collected from 26 trenches in SWDA 6. Table 8 shows the results of analyses of these samples.

The ranges of radionuclide concentrations present in these water samples are similar to those found in samples collected from wells and from seeps at the ends of trenches in SWDA 5.

Installation of Corrective Measures

Two areas have been covered with a near surface bentonite-shale seal, covering 3252 and 6875 m² as of August and November 1976, respectively (Fig. 19 and 20). Preliminary analysis of infiltrating water by means of soil-moisture resistivity blocks has been inconclusive. Quantitative evaluation of the effectiveness of the seals will include laboratory permeameter tests and field measurements with a neutron moisture-density probe.

Table 8. Radionuclide concentrations in trench monitor wells in Disposal Area $\boldsymbol{6}$

| | | | | |
|------------|-------------------|------------------------------|------------------------------|-------------------------------|
| Trench No. | Total (pCi/ml) | 90 _{Sr} (pCi/ml) | 60 _{Co} (pCi/m1) | 137 _{Cs} (pCi/ml) |
| 27 | 0.18 | 39.6 | 3.11 | <u><</u> 0.38 |
| 36 | 0.04 | 34.8 | < 0.11 | 0.23 |
| 41 | 0.14 | 15.7 | < 0.19 | <u><</u> 0.22 |
| 42 | 0.05 | 3.62 | <u><</u> 0.10 | 0.16 |
| 123 | 0.10 | 8.83 | <u><</u> 0.11 | <u><</u> 0.09 |
| 127 | 0.10 | 1.34 | <u><</u> 0.13 | <u><</u> 0.08 |
| 129 | | 43.1 | <u><</u> 0.09 | <u><</u> 0.28 |
| 135 | 0.07 | 17.4 | <u><</u> 0.13 | <u><</u> 0.08 |
| 137 | 0.13 | 7.7x10 ² | <u><</u> 0.18 | <u><</u> 0.18 |
| 139 | 0.23 | 5.23×10 ² | 0.12 | <u><</u> 0.06 |
| 145 | < 0.01 | 51.3 | ≤ 0.11 | 1.35 |
| 150 | 0.40 | 1.33 | <u><</u> 0.41 | 0.41 |
| 152 | 0.49 | 7.39x10 ² | <u><</u> 0.12 | 4.01 |
| 153 | 0.04 | 3.1x10 ² | <u><</u> 0.13 | <u><</u> 0.18 |
| 156 | 0.09 | 4.40 | <u><</u> 0.13 | <u><</u> 0.09 |
| 160 | 0.07 | 54.1 | <u><</u> 0.09 | <u><</u> 0.08 |
| 161 | 0.27 | 12.4 | <u><</u> 0.17 | 1.12 |
| 163 | <u><</u> 0.01 | 8.65 | <u><</u> 0.09 | <u><</u> 0.01 |
| 166 | 0.06 | 17.0 | 0.11 | <u><</u> 0.12 |
| 168 | <u><</u> 0.01 | 6.04 | <u><</u> 0.13 | ≤ 0.13 |
| 171 | < 0.01 | 55.9 | <u><</u> 0.13 | <u><</u> 0.39 |
| 173 | 0.13 | 8.87×10 ² | <u><</u> 0.19 | <u><</u> 0.20 |
| 177 | <u><</u> 0.01 | 30.7 | <u><</u> 0.17 | 0.79 |
| 177A | 0.18 | 8.47 | <u><</u> 0.22 | <u><</u> 0.09 |
| 279 | < 0.01 | 3.05 | <u><</u> 0.08 | <u>≤</u> 0.10 |
| 317 | 0.09 | 1.94 | <u><</u> 0.09 | <u><</u> 0.08 |

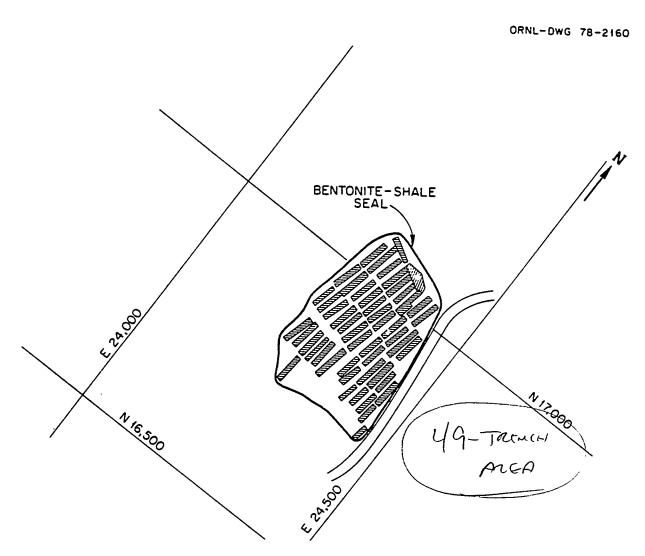


Fig. 19. Bentonite-shale surface seal in SWDA-6.

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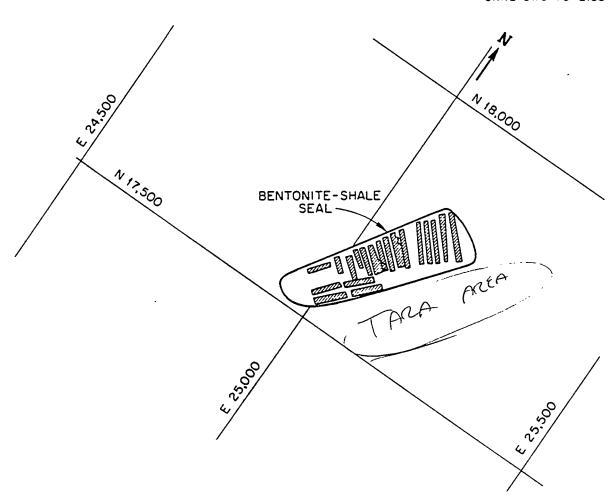


Fig. 20. Bentonite-shale surface seal in SWDA-6.

GROUND WATER TRACER STUDIES

Halocarbons as Ground-Water Tracers

During 1976 we were involved in a program of studies designed to develop the use of halocarbons as tracers of ground-water movement.

The development program was pursued through a subcontract with the Departments of Chemistry and Geology at Indiana University.

The overall objective of the program included the development of broadly applicable and highly sensitive techniques for tracing ground-water flow directions and velocities; these techniques were then applied in the Solid Waste Disposal Areas at ORNL. Unlike dyes, which are, in general, polar and of large molecular weight (molecular weights well over 200, more than 10 carbon atoms), the halocarbons investigated are nonpolar and generally contain only a single carbon atom. While they qualify as organic compounds, they could be compared to a rare gas such as xenon. Accordingly, these small, nonpolar halocarbons do not interact strongly with inorganic aquifer materials and tend to be carried along at a velocity approaching that of the ground water itself. At the same time, the uniquely high tendency of volatilized halocarbons to capture gas-phase electrons forms the basis of an exceptionally sensitive analytical technique which meets ground-water tracing requirements.

The analytical instrumentation was developed at Indiana University prior to the initiation of the subcontract. A field-operable instrument was built and tested, demonstrating that all the sensitivity obtainable in the laboratory could be attained by a field instrument

which incorporated three basic subsystems: a gas-extraction unit, in which halocarbons are stripped from the water sample by a pre-cleaned purge gas; a gas-chromatographic system, in which a network of valves and columns isolate the halocarbon of interest in a minimum period of time; and a detector system, which quantifies halocarbons down to the picogram level.

The initial phases of the tracer development program were concerned primarily with trichlorofluoromethane (CCl $_3$ F) or F-11, a compound widely known by the duPont trade name "Freon-11," which has been commercially produced in the United States since 1931. F-11 and F-12 (CCl $_2$ F $_2$) quickly became the most widely used gases in the refrigeration and air conditioning industries. By 1943, the aerosol industry began using chlorofluoromethanes as propellants in spray cans; this has become the largest single use for these compounds.

The industrial history of chlorofluoromethanes is of particular interest due to their present environmental distribution. $CC1_3F$ concentrations in ground water are typically 10^{-12} g $CC1_3F/m1$ H $_2O$ or less; the presence of F-11 in ground water is potentially significant in terms of the ground-water age. This is because the $CC1_3F$ must derive from atmospheric rain-out, and the atmospheric concentration of F-11 has increased steadily during the last 30 to 40 years.

Due to the lack of previous ${\rm CCl}_3{\rm F}$ measurements in ground water, it was necessary to investigate the basic relationship between ${\rm CCl}_3{\rm F}$ concentration and ground-water age. As a first approach to this problem, the ${\rm CCl}_3{\rm F}$ distribution was examined at sites where the

hydrology had been extensively studied, and where tritium measurements had been made in the past. Thus, the F-11 data could be evaluated by comparison with the known hydrology and inferred ground-water ages. Three areas were investigated.

The Wharton Tract, New Jersey

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The Wharton Tract is an area of 389 km² located in the Mullica River basin in the Pine Barrens region of southern New Jersey (Fig. 21). The principal aquifers in the area are the Kirkwood Formation of middle Miocene age and the Cohansey Sand Formation of Miocene and Pliocene age. In this study, all ground-water measurements made were within the Cohansey Formation, which consists of unconsolidated sediments, dominantly yellow (limonitic) quartz sand with minor amounts of gravel, silt, and interbedded clay. The site consists of a series of well clusters arranged along a line perpendicular to the river (Fig. 22). Each cluster consists of four wells screened at different depths: (1) the water table surface; (2) 7.6 m, (3) 15.2 m, and (4) 30.5 m. Clay layers occur between the 7.6-, 15.2-, and 30.5-m zones; however, confinement is not complete and a hydraulic connection exists from the water table to a depth of 30.5 m or more. A profile sketch through the "J line" (Fig. 23) shows the wells and other locations that were sampled for tritium in 1958 (Carlson et al., 1960), and for CCl₃F in 1976. The water level measured in each well during February and May, 1976, is also shown in Fig. 23.

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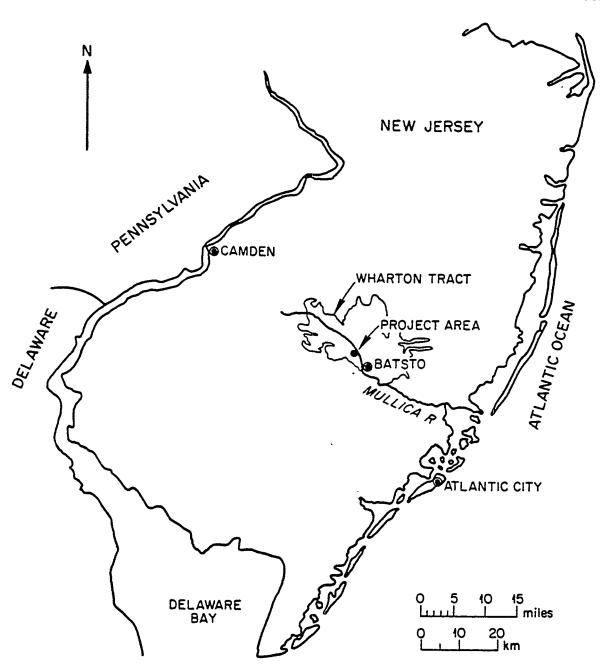


Fig. 21. Map of southern New Jersey showing the location of the Wharton Tract and project area on the Mullica River.

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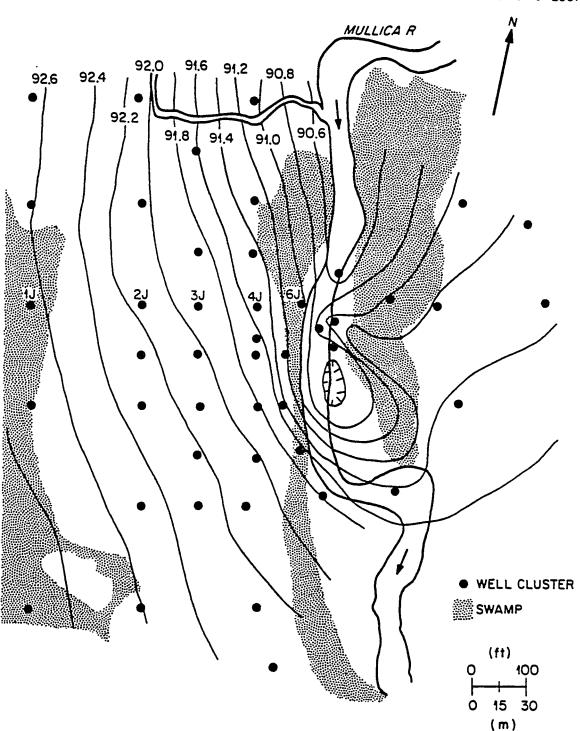
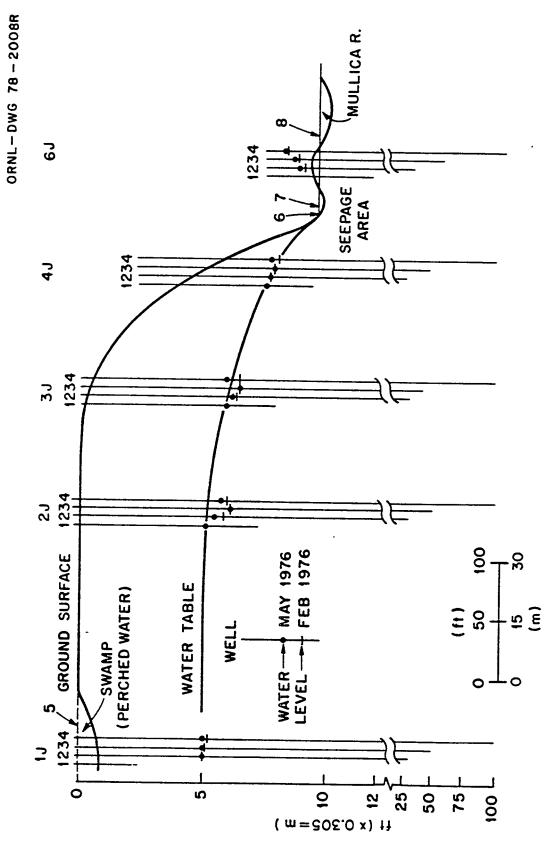


Fig. 22. Map of the study area in the Wharton Tract showing location of "J line" wells adjacent to the Mullica River.



Cross-sectional profile through the "J-line" wells showing the water table surface and the water levels in the wells during February 1976 and May 1976. 23.

The 1958 tritium measurements from Carlson et al. (1960) for the "J line" are given in Table 9. These data indicate that horizontal movement of ground water predominates even though upward movement of ground water is indicated by the piezometers near the river, and four-year-old water has reached a depth of at least 15.2 m.

In February and May, 1976, F-11 concentrations were measured in the "J line" wells, the adjacent swamps, and in the Mullica River (Table 9). After verifying that CCl_3F diminishes with depth, measurements were not made below the point where CCl_3F could no longer be detected. Although a trace of F-11 was observed in all wells, the amounts can be clearly attributed to contamination during sampling, and therefore "NDA" in Table 9 means not detectable above the background (0.005 pg/g).

The distribution of CCl₃F in the ground water at the Wharton Tract can be explained logically in terms of the hydrologic conditions. The CCl₃F concentration at the water table decreased near the river, and is attributable to dilution by older water containing no CCl₃F moving upward into the river. The absence of measurable CCL₃F below 7.6-m suggests that the majority of the water recharged locally moves along the water table surface to the river without circulating much below the 7.6-m horizon. The absence of CCl₃F in the 15.2- and 30.5-m deep wells may be due, simply, to the greater age of this water. However, the tritium study indicates considerably younger water at and below the 15.2-m level. The cause of this discrepancy is not known, but the difference could be resolved by making concurrent tritium and CCl₃F measurements.

Table 9. Tritium and CC13F measurements in the Wharton Tract

| Location No.a | Tritium (Tu)b | CC1 ₃ F (ppt) | CC1 ₃ F (ppt) |
|------------------------------|----------------------|--|--|
| | 1958 | Feb. 11-14, 1976 | May 22-25, 1976 |
| 1J-1 | 112 | 0.08, 0.06 | 0.02 |
| 1J-2 | 45 | ND ^C | ND |
| 1J-3 | 8 | ND | NA |
| 1J-4 | 2 | NA ^d | NA |
| 2J-1 | 87 | 0.40, 0.40 | O.66 |
| 2J-2 | 34 | 0.02, 0.02 | ND |
| 2J-3 | 26 | ND | ND |
| 2J-4 | 2 | ND | NA |
| 3J-1 | 108 | 0.46, 0.46 | 0.74, 0.74 |
| 3J-2 | 48 | ND | 0.07, 0.09, 0.09 |
| 3J-3 | 31 | NA | ND |
| 3J-4 | 0 | NA | NA |
| 4J-1 4J-2 4J-3 4J-4 | 131 35 22 0 | ND NA NA | 0.11, 0.08 ND NA NA |
| 6J-1 | 147 | 0.07, 0.07 | ND |
| 6J-2 | 21 | ND | ND |
| 6J-3 | 8 | NA | NA |
| 6J-4 | 0.8 | NA | NA |
| 5 6 7 8 | | 0.91, 0.90 (3.7°C) 0.11 (7.5°C) 0.29 (7.5°C) 0.91 (3.7°C) | 0.01 0.21, 0.34 (17°C) 0.48, 0.44, 0.40 (20° |

aNumbers refer to Figure 22.

bCarlson et al. 1960.

cNot detectable < 0.01 ppt.</pre>

dNot analyzed.

Hot Springs, Arkansas

Two springs in the Hot Springs National Park were tested for CCl_3F content (Table 10). The age of the water released from these springs is known from extensive hydrologic and geochemical study. Tritium and ^{14}C analyses of the waters indicate that the springs yield a mixture of a small amount of water less than 20 years old, with a preponderance of water about 4400 years old (Bedinger et al., 1974).

The tritium content of the two springs tested (Table 10) is very low, less than 3 TU in both cases, and is attributed to mixing of the dominant older water with a small amount of recent water (less than 1%) near the spring exit. Values for F-11 in these springs were also very low, both less than 0.003 ppt. Using 0.003 ppt as a maximum spring value, and a representative value for recent surface water (0.45 ppt), a maximum mixing ratio of 0.7% is indicated. In this regard, the F-11 results agree very well with the tritium data.

Edwards Aquifer, Texas

A sampling program was undertaken in the Edwards aquifer in the San Antonio, Texas, area to determine whether a correlation exists between the CCl $_3$ F distribution and recent tritium measurements. The study revealed a series of anomalous CCl $_3$ F concentrations (35 ng/ml to 2 ng/ml H $_2$ O) that are too high to be of atmospheric origin. The anomalous points occurred in a line extending from the northwest corner of San Antonio for a distance of 74 km northeast along the Balcones fault zone. The location of the plume indicates direction of ground-water movement, and demonstrates the likely utility of CCl $_3$ F

Table 10. Tritium and CCl₃F measurements from Hot Springs National Park, Arkansas

| USGS No. | Tritium (TU ± 1σ) | F-11 (ppt) | Comments |
|-----------------|-------------------|------------|-----------------------------|
| 343110093025301 | 1.1 ± 0.6 | 0.003 | Happy Hollow Spring |
| 343052093050802 | 2.8 ± 0.5 | 0.003 | Wittington Avenue Spring |

as a ground-water tracer. Away from the plume, most ambient ${\rm CCl}_3F$ levels correlated well with tritium abundances. This correlation is important because it suggests that ambient F-11 can be used as a tool to study ground water in much the same manner that tritium has been used.

Laboratory Measurements of Tracer Mobility

From a practical point of view, the question about any tracer is the rate at which it is capable of moving and, in particular, the relationship between the velocity of the tracer (u_t) and the velocity of the ground water (u_w) . The ratio u_t/u_w is commonly referred to as the <u>retardation factor</u> in chromatographic studies. Treating the migration of tracers in natural porous media essentially as an experiment in chromatography, the retardation factor (R) can be measured directly as:

$$R = \frac{u_t}{u_w} = \frac{V_w}{V_R} \quad ,$$

where, for a chromatographic column packed with some porous medium, $V_{\rm w}$ is the volume of the aqueous or mobile phase, and $V_{\rm R}$ is the retention volume or breakthrough volume of the tracer.

The magnitude of $V_{\rm W}$ is determined primarily by the porosity of the natural medium, more specifically by the pore-volume occupied by water which is actually mobile and participating in the flowing system. Quantitatively, the magnitude of $V_{\rm W}$ is given directly by the retention volume of any unretained component. For many natural media,

chloride ion can be safely taken to be an unretained species, and the retardation factors given here are all based on the assumption that, for the sand and limestone columns studied, $V_{\rm W}$ is given directly by the retention volume of chloride ion. Obtaining a precise value for R depends on minimizing the uncertainties in $V_{\rm W}$ and $V_{\rm R}$, a goal which can be met by minimizing the purely physical dispersion processes that tend to make solute-zone boundaries indistinct.

Striving to make the best possible measurements of R values for potential tracers, we have considered reliable accuracy and high precision to be the most important qualitities of any practical system. We have developed and used three different systems within the past year, the sequence representing a very substantial evolution. A fourth system is to be constructed in the coming year and is described here for completeness. All four systems are summarized in Table 11.

The results of many different column tests for a large number of potential halocarbon tracers are summarized in Tables 12 and 13. It can be noted that significant retardation has been observed only for the compounds C_8F_{16} and $CCIF_2CICIF$. We regard these results as secure within the uncertainties quoted and suggest that they represent a useful beginning for an interesting series of tests dealing with the mobilities of these same substances on progressively more active (i.e., retentive) natural media. There is every reason to believe that extensive further results can be obtained.

Table 11. Comparison of methods used for the determination of R (Retardation factor)

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| | System I | System 11 | System III | System IV |
|--|--|---|---|--|
| Method of column development | Elution of spike | Frontal analysis (miscible displacement) | Elution of spike | (to be constructed) Frontal analysis (miscible displacement) |
| Column Dimensions Particle size Applied pressure Typical V, Efficiency | 10 mm x < 1 m 100 < d < 150 µm < 0.05 atm 50 ml 20-50 theoretical plates | Same as I | (sand) (limestone) 4.0 mm x 1.50 m 4.0 mm x 50 cm 74-150 μm | 4.0 mm × 50 cm 10-20 µ 50-100 atm > 5000 |
| Sampling Uncertainty in VR | Discontinuous, 1-3 ml aliquots ~ ± 3 ml | Continuous monitoring ± 1 mi | Continuous monitoring <u>via</u> continuous gas-stripping of effluent ± 0.3 ml | Continuous monitoring |
| Method Nature of compounds which can be analyzed | Electron capture gas chromatography Electron-capturing species exhibiting high volatility | Mass spectrometry <u>via</u> permeable membrane Any species transmitted by membrane | Nonspecific measurement of all volatile electron-capturing species Same as I | E 7:0 A |
| Selectivity Number of compounds which can be analyzed | Excellent Depends on gc performance, usually 5-6 | Excellent Depends on spectra, usually 10-12 | Specific determination of more than one electron-capturing compound requires resolution on column | Same as 11 |
| Minimum detectable tracer concentration Time required | 1011 <u>M</u> < 10 hr | 10-7 <u>M</u> < 10 hr | 10-10 <u>H</u> << 20 min | |

^aValue given for V_W includes substantial contribution by dead volume in injector system. The dead volume also contributes substantially to solute-zone broadening and the true column efficiency is therefore not known.

Table 12. Retardation factors of potential tracers in sand

| | System I | | Syste | em II | Syste | em III. |
|---|---------------------|------------|--|-------------------------------------|--|---|
| | V _R , ml | R | V _R , ml | R | V _R , ml | R |
| C1- SF ₆ Ar | 50 ± 3 | ≡ 1.0 | 45.5 ± 1.2 | ≡ 1.0 | 15.8 ± 0.3 15.6 ± 0.3 | ≣ 1.0 1.01 ± 0.05 |
| AI . | | | 45.6 ± 1.2 | 1.0 ± 0.1 | | |
| CH2C12 CHC13 CC14 CH3CC13 CH3CH2CC13 | | | 43.8 ± 1.6 44.3 ± 1.6 47.5 ± 1.0 | 1.0 ± 0.1 1.0 ± 0.1 1.0 ± 0.1 | 16.0 ± 0.3 16.0 ± 0.3 16.0 ± 0.3 15.8 ± 0.3 15.8 ± 0.3 | 0.99 ± 0.05 0.99 ± 0.05 0.99 ± 0.05 1.00 ± 0.05 1.00 ± 0.05 |
| CHC1 ₂ F CC1 ₂ F ₂ CC1 ₃ F | 50 ± 3 | 1.0 ± 0.1 | 45.8 ± 1.2 | 1.0 ± 0.1 | 15.8 ± 0.3 15.7 ± 0.3 15.7 ± 0.3 | 1.00 ± 0.05 1.01 ± 0.05 1.01 ± 0.05 |
| CBr ₂ F ₂ CBr ₃ F | 50 ± 3 | 1.0 ± 0.1 | 46.0 ± 1.2 45.6 ± 1.2 | 1.0 ± 0.1 1.0 ± 0.1 | 15.7 ± 0.3 | 1.01 ± 0.05 |
| CC1F2CC1F2 CC12FCC1F2 CC13CF3 CC12FCC12F | | | 47.0 ± 1.6 46.6 ± 1.2 47.0 ± 1.6 | 1.0 ± 0.1 1.0 ± 0.1 1.0 ± 0.1 | 15.8 ± 0.3 16.0 ± 0.3 | 1.00 ± 0.05 0.99 ± 0.05 |
| CBrF ₂ CBrF ₂ CC1F ₂ CIC1 CI ₂ FCF ₃ | | | 45.8 ± 1.2 46.2 ± 1.2 48.0 ± 1.6 | 1.0 ± 0.1 1.0 ± 0.1 0.9 ± 0.1 | | |
| C8F16 | 200 ± 20 | 0.25 ± 0.2 | | bserved | not ob: | served |

Table 13. Retardation factors of potential tracers in limestone

| Compound | Sys | tem I | Syste | em II |
|--|----------|---------------|--|-------------------------------------|
| | VR, ml | R | VR, ml | R |
| C1- N ₂ | 46.2 ± 2 | ≡ 1.0 | 46.8 ± 1.6 51.1 ± 1.4 | ≡ 1.0 0.9 ± 0.1 |
| CH ₂ C1 ₂ CHC1 ₃ CC1 ₄ | | | 45.5 ± 1.6 47.8 ± 1.6 49.0 ± 2.0 | 1.0 ± 0.1 1.0 ± 0.1 1.0 ± 0.1 |
| CC1 ₃ F | 50 ± 2 | 0.9 ± 0.1 | 49.0 ± 2.0 | 1.0 ± 0.1 |
| CBr ₂ F ₂ CBr ₃ F | . 50 ± 2 | 0.9 ± 0.1 | 50.0 ± 1.8 48.5 ± 1.3 | 0.9 ± 0.1 1.0 ± 0.1 |
| CC1 ₂ FCC1 ₂ F | | | 48.4 ± 1.4 52.6 ± 1.4 | 1.0 ± 0.1 0.9 ± 0.1 |
| CBrF ₂ CBrF ₂ CC1F ₂ CIC1F | | | 47.8 ± 1.4 55.9 ± 1.4 | 1.0 ± 0.1 0.8 ± 0.1 |

622 Mean 313

Tracer Test at Solid Waste Disposal Area 6

A ground-water tracing test using dibromodifluoromethane (CBr_2F_2) was carried out adjacent to SWDA 6. This test represents the first practical application of the idea that halocarbons will be useful as ground-water tracers.

The test site consists of one of several arrays of wells installed by ORNL to obtain information about ground-water movement adjacent to the various disposal areas. Tracing tests using tritium are currently being conducted at these sites, but at the time of the CBr_2F_2 test no successful tracing test had yet been performed. Because no information was available regarding important parameters such as tracer dilution and direction of ground-water flow, these factors had to be roughly estimated. Accordingly, the CBr_2F_2 test was geared to produce only approximate information that would serve principally as a guideline for planning a subsequent tracing test at the same site.

The test site is in the vicinity of SWDA 6 adjacent to White Oak Creek in an area where the ground water is uncontaminated by radioisotopes or other waste. The aquifer in the test area is the Conasauga Shale, which possesses primarily fracture permeability. Seven observation wells are arranged in a semi-circle on a 9-m radius around a central injection well (Fig. 24).

The tracer solution was prepared by mixing 23 mg $\mathrm{CBr}_2\mathrm{F}_2$ with 5 ml ethanol. This solution was then mixed with one liter of water and injected into the central well. The sampling schedule was based on expectations that the tracer would require one month or longer to reach the observation wells. Therefore, sampling on weekly intervals,

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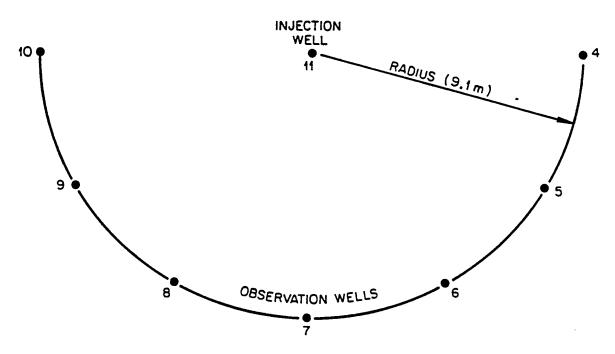


Fig. 24. Well layout and numbering system.

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beginning two weeks after the injection, seemed appropriate for this preliminary type of tracer test. After eight sampling periods the accumulated water samples were mailed to the University of Arizona where they were analyzed on a Varian Model 3700 gas chromatograph.

The analytical results are presented in Table 14 and shown graphically as a function of time in Fig. 25. Anomalously high tracer concentrations appeared in well 6. The adjacent wells, 5 and 7, also showed values that were slightly above the background (about 0.02 ppt for this test), but no anomalous tracer concentrations were detected in any of the other observation wells.

The results indicate quite conclusively that the tracer was intercepted in well 6. This well, which is directly down gradient from the injection point, showed the earlier anomalous tracer concentration and is the well in which the highest values were observed.

Furthermore, the shape of the tracer concentration versus time plot observed in well 6 (Fig. 25) compares very favorably with the tracer breakthrough curve that would be expected on a theoretical basis for this type of test. These observations, as well as the fact that a reasonable ground-water velocity is indicated from the arrival time of the peak concentration in well 6, constitute strong evidence that the tracer was intercepted.

The average ground-water velocity for a test of this type, in which the tracer is injected as a single pulse, is theoretically determined from the arrival time of the peak tracer concentration in the down-gradient observation well. In this test the highest

Table 14. CBr_2F_2 concentration in each well (ppt)

| Complian. | Well Number | | | | | | | | |
|-----------------|-------------|------|------|------|------|------|-------|-----|--|
| Sampling period | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | |
| 1st | 0.01 | 0.02 | 0.28 | 0.03 | 0.03 | 0.01 | 0.01 | 754 | |
| 2nd | 0.03 | 0.11 | 0.40 | 0.08 | 0.02 | 0.01 | 0.000 | 141 | |
| 3rd | 0.00 | 0.02 | 0.18 | 0.06 | 0.01 | 0.01 | 0.01 | 11 | |
| 4th | 0.01 | 0.12 | 0.09 | 0.09 | 0.03 | 0.01 | 0.00 | 1.7 | |
| 5th | 0.01 | 0.03 | 0.05 | 0.08 | 0.01 | 0.01 | 0.00 | 1.5 | |
| 6th | 0.00 | 0.01 | 0.06 | 0.06 | 0.2 | 0.01 | 0.01 | 2.1 | |
| 7th | 0.02 | 0.02 | 0.03 | 0.05 | 0.03 | 0.01 | 0.01 | 1.5 | |
| 8th | 0.02 | 0.00 | 0.03 | 0.06 | 0.04 | 0.03 | 0.04 | 1.3 | |

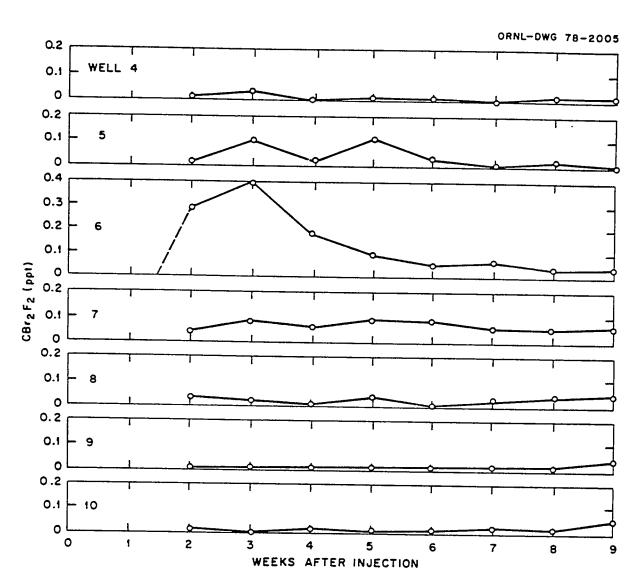


Fig. 25. Tracer concentration in each well versus time.

concentration was observed in well 6, 23 days after the injection. Because the wells were sampled only at weekly intervals, however, the possible error in this observation may be as much as \pm 3.5 days (\pm 15%). Considering that the distance traveled was 9.1 m, the minimum velocity indicated is 0.4 \pm 0.06 m/day. Although the distribution coefficient for CBr₂F₂ on Conasauga Shale has not been determined, column tests have shown that CBr₂F₂, when introduced at a concentration of 1 mg/g, moves 0.71 times as fast as the water through a permeable column of quartz sand. If this scale factor is applied to the tracer test data, the interpreted ground-water velocity becomes 0.6 \pm 0.1 m/day.

From the velocities determined from the tracing test and the rate of tracer dilution observed in the injection well, a range of values for the effective porosity of the Conasauga Shale can be calculated. The measured velocity extremes, 0.4 m/day and 0.5 m/day, correspond to effective porosities of 3% and 2%, respectively. Considering that typical effective porosity values for fractured shales at shallow or moderate depths normally range between 0.5% and 5%, the values calculated from the dilution data appear to be quite plausible.

Although this test was performed to obtain information about ground-water flow in the vicinity of SWDA 6, the test also served as a means of evaluating the tracing technique itself. The fact that relatively unambiguous results were obtained on the first attempt at a test site where no successful tracing test had been performed previously is proof that the technique is quite viable and should, therefore, be developed further.

CORRECTIVE MEASURES

The transport of radionuclides from the disposal areas is primarily a function of the amount of infiltration of rainwater, the amount of leaching of the waste by water, and the amount of leachate leaving the waste trench as either ground-water flow or surface seepage. The corrective measures discussed in the following sections address the problem of reducing the amount of water that comes in contact with the buried waste.

Design of the Bentonite-Shale Mixture

To impede the infiltration of ground water into the radioactive solid waste trenches, a near-surface seal of bentonite (a swelling clay) and shale mixture is being developed. To function properly as a seal, the admixture must contain enough bentonite to fill the voids in the shale matrix. Maximum reduction in permeability is obtained when the seal is compacted to a maximum density at an optimal moisture content.

A series of laboratory tests were performed to determine the optimal bentonite/shale composition of the admixture and the optimal moisture content to obtain maximum compacted densities. Compaction curves, as shown in Fig. 26, were compiled for three bentonite/shale percentages. Ten-centimeter-diameter compaction molds were used in the standard Proctor tests as described by Lambe (1960). Maximum densities are obtained in compacting the admixture at approximately 20% moisture content during field installation. Higher percentage bentonite admixtures expectedly yield lower compacted densities due to the

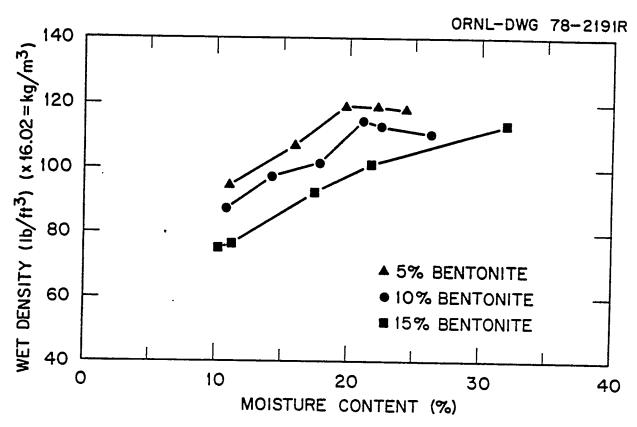


Fig. 26. Optimal moisture content compaction curves for the three bentonite-shale admixtures.

relative bulk density of the bentonite. Direct measurement of the effectiveness of the bentonite/shale mixture will be obtained in laboratory permeameter tests at a constant hydraulic head.

Expansion Test

To more fully evaluate the effectiveness of the bentonite-shale surface seals in the disposal areas, a laboratory study of the expansive property of the admixture was performed. Compaction molds (10-cm dia.) were filled with bentonite mixtures (triplicate molds at 10 and 15% bentonite by weight, and at 20% moisture content) by the Standard Proctor method. Degree of expansion of the admixture exposed to water at the base of the mold was measured over a three-week period.

Several tests were performed with the admixture under varying confining stress in order to determine the effect of replacement of 0.6 m of overburden over the seal. For a 10% bentonite admixture a 6.5% increase in volume was observed after seven days when unconfined (Fig. 27, solid line); whereas, only a 2.5% increase was observed after three days when subject to the equivalence of 0.37-m overburden (dashed line). For a 15% bentonite admixture subject to confining overburden (dot-dash line), more rapid expansion to an ultimate increase 1.5 times greater was expectedly observed. Correlation of the rapidity and degree of expansion indirectly determines the effectiveness in permeability reduction.

From such laboratory tests several conclusions about bentonite-shale admixtures and seal installation procedures can be made. Most important is that the bentonite application must be

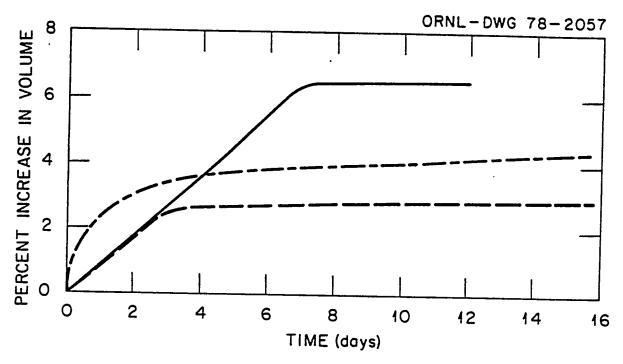


Fig. 27. Bentonite-shale admixture expansion tests (10% bentonite, unconfined solid line; 10% bentonite, 0.37 m overburden, dashed line; 15% bentonite, 0.37 m overburden, dot-dash line).

sectionally completed over a small area at a time. Mandated by the rapidity of expansion of the seal, compaction of the seal by rolling and replacement of the shale overburden must be completed without the interference of rainfall. If allowed to expand prior to replacement of the shale overburden, the bentonite seal will not attain the required permeability reduction.

Inherent in this laboratory study is the assumption that maximum compacted densities are obtained by rolling the seal at optimal moisture content. A sufficient volume of water and associated spraying apparatus will be required for this operation. Alteration of field application will void the results of this idealized study.

Associated with a higher percentage bentonite admixture is a more rapid expansion and greater ultimate increase in volume. Field application at these higher rates mandate more stringent requirements of maintaining optimal moisture contents during compaction and immediate replacement of shale overburden.

Although of a preliminary nature, this study of the expansive property of the seal aids in the design of future bentonite applications in attaining the required permeability reduction. Further physical and chemical laboratory analyses, in conjunction with field studies, will be necessary to fully evaluate the use of bentonite admixtures as surface seals.

GROUND-WATER TRANSPORT MODELS

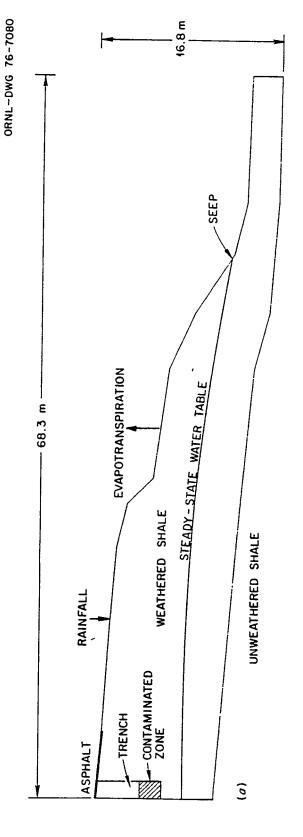
Numerical modeling of contaminant transport in ground-water systems is an essential aspect of the disposal area studies. Recently

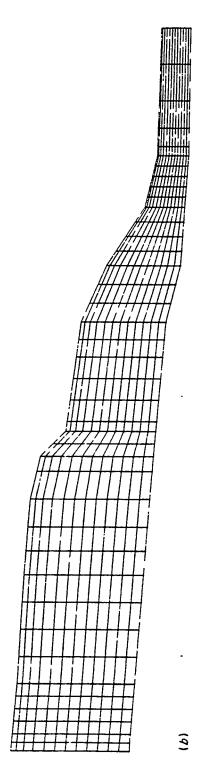
developed models can be applied in the prediction of radionuclide transport from existing and proposed disposal areas, evaluation of an accidental release at power reactor sites, and investigation of proposed methods for decreasing radionuclide transport from the disposal area.

A two-dimensional computer code for simulation of radionuclides in a combined saturated-unsaturated flow regime has been developed (Reeves and Duguid, 1975a and b; Duguid and Reeves, 1975). The mathematical formulation incorporates mechanisms for the convective transport, hydrodynamic dispersion, chemical adsorption, and radioactive decay. The Galerkin finite element formulation utilizes bilinear interpolation for the spacial discretization of the porous media.

The model has been applied to the transport of tritium at the intermediate-level liquid waste trench 7 at ORNL in order to determine whether a simplified rainfall boundary condition can be used. A cross section of the seepage trench and its associated surface seep is shown in Fig. 28a. The transporting fluid (water) enters the unweathered shale in the form of infiltrating precipitation, travels down toward the water table, and eventually emanates at the surface seep. A transient rainfall boundary allows an evapotranspiration flux out of the shale. In this application, radionuclide transport through the unsaturated zone is an important consideration.

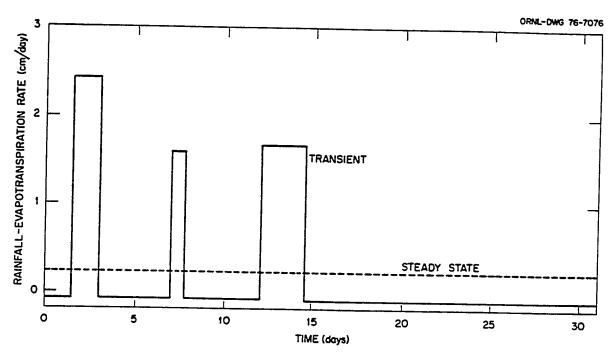
To evaluate the sensitivity of the model to rainfall flux boundary condition along the soil surface, two cases were examined. One boundary condition, labeled "steady-state" in Fig. 29, represents the net precipitation of 77.7 cm/year (average rainfall less





(a) Cross section through seepage trench 7 showing the relationship of the trench to a nearby surface seep, the asphalt covering above the trench, and the steady-state water table. (b) Finite-element discretization of the region for simulation of water movement and tritium transport. Fig. 28.

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Rainfall boundary conditions for the month of May 1972. The steady-state condition represents the average precipitation less the average evapotranspiration. The transient condition shows three storms superposed on the average evapotranspiration. Fig. 29.

evapotranspiration), as uniformly distributed over a one-month time period. The "transient" boundary condition was developed from precipitation data for May 1972 such that both conditions would yield the same integrated infiltration for the month.

Numerical solution of the soil-water transport equation provided the Darcian velocities necessary to simulate the radionuclide transport for both cases. The adsorption of tritium was assumed to be zero (Kd = 0). Published values of dispersivities were used, as field-measured values for the Conasauga Shale are not yet available.

The simulation of the movement of the isopleths in Fig. 30 show that even though the trench is covered with an asphalt seal, lateral movement of soil water in both the saturated and unsaturated zones provides radionuclide transport from the buried waste. The effect of individual rain storms on radionuclide transport was shown to be negligible, thus average rainfall conditions may be applied in future considerations. This observation drastically reduces the computer execution time and core requirements for transport simulation.

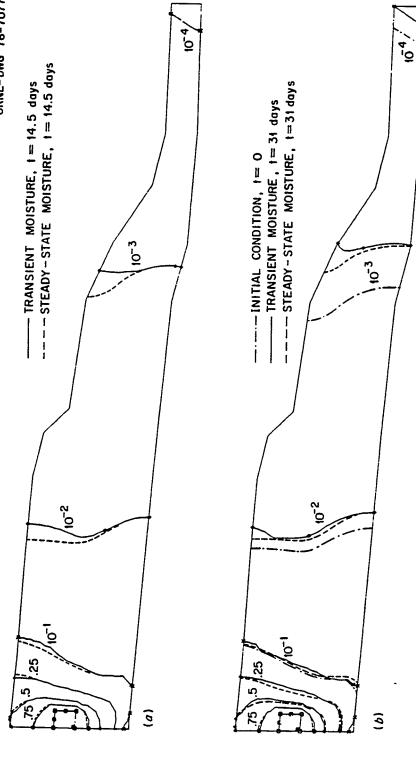
Unrealistic amounts of radionuclide transport were observed in the unsaturated zone above the water table. Attributed to the high values of dispersivity used in the model, radionuclide transport in unsaturated soils is an area of further research.

SHALLOW LAND BURIAL DATA BASE

The staff of the Ecological Sciences Information Center (ESIC) of the Information Center Complex has been compiling an information file in collaboration with Dames and Moore of White Plains, New York, to



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Concentration contours resulting from both steady-state and transient-state rainfall conditions at (a) $t=14.5\,$ days and (b) $t=31\,$ days. The initial (t=0) condition is also given in (b). Fig. 30.

provide information support to the Department of Energy/Energy

Technology, Division of Waste Management and its contractors. Priority

is being placed on obtaining abstracting, and incorporating documents

which deal with the DOE "old" Manhattan sites, commercial disposal

sites, and the specific parameters which affect the soil and geologic

migration of radionuclides. Additional literature selected for

inclusion in the data base deals with container design and performance,

waste production and processing, legal and political aspects, and

nonnuclear waste disposal.

Each reference in the Shallow Land Burial Data Base contains a complete bibliographic citation, an informative abstract emphasizing the important findings, subject categories describing the general subject area, keywords identifying specific topics in the document, and data and location of data-taking activities. Specialized data fields have been added to improve the ease and accuracy of identifying pertinent references. Specific radionuclides for which data are presented are listed in a field called "Measured Radionuclides." When information is presented on parameters which affect the migration of these radionuclides through the soil and geological formations, the parameters discussed are listed in a field entitled "Measured Parameters." These parameters are also discussed in the abstract along with their values, where possible. These fields enable the user to review only those references to documents concerned with the radionuclides and parameters of interest.

Each document referenced in the Shallow Land Burial Data Base is assigned a "relevance" number which is recorded in a special field

called "Data Index." The number facilitates sorting documents according to their pertinence to shallow land burial technology.

Documents are rated 1, 2, 3, or 4, with 1 indicating direct applicability to shallow land burial technology and 4 indicating that a considerable amount of interpretation is required for the information presented to be applied. The "relevance" number does not reflect the quality of the document, only its applicability to shallow land burial technology.

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